

Sampling PM_{2.5} emissions from coal combustion: Effects of dilution ratio and residence time

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Summary

Particulate matter (PM) is emitted directly from sources (primary PM) and formed in the atmosphere from reactions of gaseous precursors (secondary PM). Coal-fired utility boilers are a source for both primary PM and gaseous precursors that react in the atmosphere to create secondary PM. This research examines the size distribution and chemical composition of primary PM emissions from coal combustion.

Primary PM emissions from coal combustion typically have a bimodal distribution. The majority of the PM mass occurs in the coarse mode (particles larger than 1 micrometer). On a number basis, the majority of the PM occurs in the submicron mode with a mean diameter around 0.1 micrometer; these particles are formed by vaporization, condensation, and nucleation of inorganic constituents in the fuel. The effectiveness of air pollution control devices such as bag houses and electrostatic precipitators at removing particles is size dependent. These devices effectively remove coarse mode particles, but are less effective at controlling submicron particles. The exact size distribution of the PM emissions from a coal-fired power plant depends on the coal quality, combustion technology, and the effectiveness of the air pollution control system.

Dilution sampling is a technique that has been developed to examine the influence of rapid cooling and dilution on PM emissions from combustion systems. A dilution sampler rapidly mixes hot exhaust gases with a specified amount of conditioned air and allows for processes such as nucleation, condensation, and coagulation to occur. Although a dilution sampler cannot simulate the complexity of actual plume mixing, it allows systematic examination of the effects of dilution on PM emissions in order to better understand the PM transformations that occur in plumes.

A dilution sampler was designed and built at Carnegie Mellon University to examine the effects sampling conditions on PM_{2.5} emissions from coal boilers. The design is based on the Caltech dilution sampler, with improvements that allow for the independent control of dilution ratio and residence time to allow investigation of the effects of these parameters on emissions. This sampler was installed upstream and downstream of the bag house of a pilot-scale pulverized coal combustor. Measurements of particle size distributions from 0.003 μm – 20 μm were made for a range of dilution ratios and residence times. Filter samples of PM_{2.5} were also collected for gravimetric and chemical composition analysis. The data are compared to predictions from coagulation theory to investigate the influence of aerosol processes on the particle size distribution.

The particle size distribution measured before the bag house contained an ultrafine mode with a peak around 0.01 μm . The particles in this mode dominate the emissions of particle number, but contribute negligibly to the emissions of particle volume (and mass). Changing the dilution ratio also alters the size distribution of the submicron particles. Increasing the dilution ratio increases the particle number, especially in the ultrafine 0.01- μm mode. Changing the dilution ratio does not affect the size distribution of particles greater than 1 μm . The ultrafine mode is not observed after the bag house.

The dilution ratio and residence time do not influence PM_{2.5} mass emission rate, indicating that there is no significant transfer of mass via nucleation or condensation occurring within the residence time tank. This is not unexpected because of the low levels of volatile species in coal combustion exhaust. Dilution ratio and residence time both have a significant impact on the

total number of particles emitted. Increasing the residence time reduces the number concentration while increasing the dilution ratio increases the total number concentration.

A coagulation model was used to simulate the changes over time of the particle size distribution within the residence time tank to better understand the effects of dilution and residence time on the measured size distributions. The model was used to simulate the changes in the particle size distribution as the sample passes through the residence time tank. The model only accounts for effects of coagulation on the aerosol size distribution due to Brownian motion. The input for the model is the size distribution measured at the end of the dilution tunnel. The agreement between measured size distributions and distributions predicted by the coagulation model suggest that the main process occurring within the residence time tank for these conditions is Brownian coagulation of the emitted particles.

However, coagulation alone is not sufficient to explain the effects of dilution ratio on the size distribution. Upon entering the dilution tunnel, the combustion products are rapidly diluted and cooled causing SO_3 to react with H_2O to create H_2SO_4 , sulfuric acid. The rapid cooling of the combustion products creates a supersaturation of sulfuric acid. Whether or not this material condenses or nucleates to create new ultrafine particles depends on the available particle surface area. The lower the dilution ratio the greater amount of surface area per unit volume and the more likely that the sulfuric acid will condense onto existing particles reducing the production by nucleation of new particles in the ultrafine mode. High dilution ratios reduce the amount of available surface area per unit volume, which increases nucleation and the number of particles in the ultrafine mode. Therefore, one must account for coagulation, nucleation, and condensation when considering the effects of dilution ratio on the particle size distribution.

Filter samples were also collected for chemical analysis of trace metals. These samples were collected at different dilution ratios and residence times to examine the impact of the dilution process on the $\text{PM}_{2.5}$ chemical composition.